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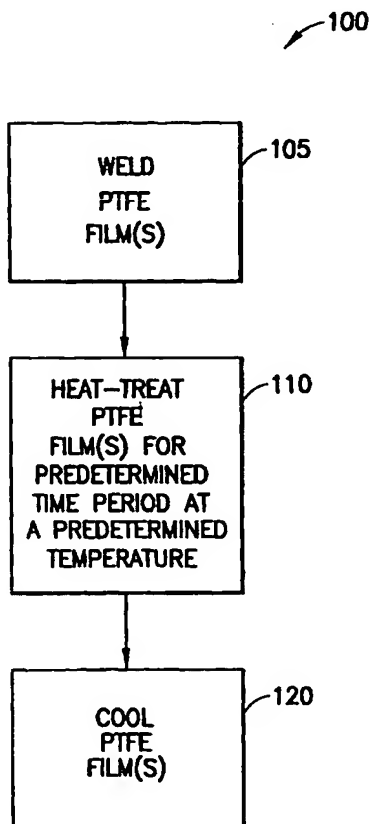
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- (21) International Application Number: **PCT/US2004/020024** (74) Agent: **WILLIAM F. RYANN**; Advanced Technology Materials, Inc., 7 Commerce Drive, Danbury, Connecticut 06810 (US).
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[Continued on next page]

(54) Title: **POLYTETRAFLUOROETHYLENE TREATMENT**



(57) Abstract: One or more PTFE films are heated to greater than 150 degrees centigrade (C) and for a time greater than 20 hours, then the PTFE films are cooled. The PTFE films may be heated to temperatures greater than 200°C and less than 250°C and most preferably heated to a temperature of about 228°C. The PTFE films may be kept at a temperature for greater than 50 hours or most preferably kept at a temperature for around 100 hours. The PTFE films may be heat processable PTFE fluoropolymer films and may have a number of heat affected zones. The heat affected zones may be created before or after heat treating. The heat affected zones are generally caused by welding two or more PTFE films together, usually under pressure. An "optimal" temperature and "optimal" time period are determined at which heat processed polytetrafluoroethylene (PTFE) fluoropolymers should be heat treated.



ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),  
European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI,  
FR, GB, GR, HU, IE, IT, LU, MC, NL, PL, PT, RO, SE, SI,  
SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ,  
GW, ML, MR, NE, SN, TD, TG).

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— with international search report

— with amended claims

Date of publication of the amended claims: 3 February 2005

*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

**AMENDED CLAIMS**

**[Received by the International Bureau on 21 December 2004 (21.12.04) ;  
original claims 1-18 replaced by amended claims 1-20]**

1. A method comprising:  
  
heating a polytetrafluoroethylene material to an elevated temperature; and  
maintaining said heating for a time sufficient to substantially reduce a  
particle count character of the polytetrafluoroethylene material.
2. The method of claim 1 further comprising applying a melting temperature to  
a portion of the polytetrafluoroethylene material for welding thereof prior to  
said heating.
3. The method of claim 2 wherein the melting temperature is within about  
15°C of a melting point of the polytetrafluoroethylene material.
4. The method of claim 2 wherein said applying forms a heat affected zone of  
the portion, said heating and said maintaining to affect the heat affected  
zone.
5. The method of claim 1 wherein the elevated temperature is above a glass  
transition temperature of the polytetrafluoroethylene material.
6. The method of claim 1 wherein the elevated temperature is between about  
130°C and about 260°C.
7. The method of claim 1 wherein the time is between about 20 hours and  
about 100 hours.
8. The method of claim 1 wherein said maintaining occurs in a periodic  
manner comprising:  
  
cooling the polytetrafluoroethylene material; and reheating the  
polytetrafluoroethylene material.

9. The method of claim 1 wherein said maintaining occurs in a periodic manner comprising:  
cooling the polytetrafluoroethylene material; and  
reheating the polytetrafluoroethylene material.
10. A method comprising heating a polytetrafluoroethylene material to about 228°C for a sufficient time to substantially reduce a particle count character thereof.
11. The method of claim 10 wherein said heating is for about 100 hours.
12. A polytetrafluoroethylene material having a particle count character reduced by application of an elevated temperature thereto.
13. The polytetrafluoroethylene material of claim 12 wherein the elevated temperature is between about 130° and about 260°C.
14. The polytetrafluoroethylene material of claim 12 wherein the application of the elevated temperature is for between about 20 hours and about 100 hours.
15. The polytetrafluoroethylene material of claim 12 wherein the elevated temperature is about 228°C and the application is for about 100 hours.
16. The polytetrafluoroethylene material of claim 12 in the form of a film for contacting a substance.
17. The polytetrafluoroethylene material of claim 16 wherein the substance is one of a liquid and a powder.
18. The polytetrafluoroethylene material of claim 17 wherein the film is incorporated a package to contain the substance.

19. A method of determining a temperature and time at which heat processible polytetrafluoroethylene (PTFE) fluoropolymer films are heat-treatable to reduce particle count thereof, comprising:

providing a set of heat processible PTFE fluoropolymer films;

subjecting each film of said set of heat processible PTFE fluoropolymer films to a predetermined temperature for a predetermined time of heat processing, wherein temperature and time of heat processing are varied among films in said set, to provide a range of heat processing temperatures and a range of heat processing times for heat processed films in said set;

after said heat processing, determining particle counts for the heat processed films in said set;

performing regression analysis on the particle counts to determine temperature and heat processing time at which particle count is minimized, as said determined temperature and time.

20. A method of treating a polytetrafluoroethylene film to reduce particle count character thereof, comprising exposing the film to a temperature in a range of from about 150°C to about 250°C for a time greater than 20 hours, sufficient to reduce particle count of said film to below 10 particles/ml of particles having a diameter of 0.2 micron.

standard deviation of those particle counts. That is, such PTFE material may be referred to as having a high particle count character. Embodiments of the present invention may be employed to reduce the particle count character of PTFE material. In fact, embodiments described herein may reduce the particle count character and the standard deviation thereof by factors greater than about ten. It should be noted that particle counting systems may also count "microbubbles" or other anomalies as "particles" accounted for in establishing a particle count or the particle count character of a material. The term "particle," as used herein, is intended to encompass whatever can be construed to be a "particle" by a conventional particle counting system.

[0023] Referring now to FIG. 1, a method 100 is shown for heat treating PTFE films in order to reduce particulate shedding. Method 100 may be employed where PTFE films are to be placed in contact with liquid substances. The method 100 may be used whenever particle count reduction is desirable. This may also include situations where PTFE films are to contact non-liquids such as substances.

[0024] In step 105, a PTFE film is welded. This step is optional. In general, however, most liners made of PTFE will be welded. Welding for PTFE films is a process that melts and joins separate PTFE film portions generally under an amount of pressure. As described in more detail below, welding occurs at about the melting point of the particular PTFE film employed.

[0025] Accordingly, in one embodiment, welding may include exposing the PTFE films to between about 370C and about 430C for between about 5 and about 15 seconds. A pressure of between about 60 psi and about 100 psi may be applied. Additionally, the PTFE films may be cooled for between about 5 and about 15 seconds at a constant pressure before further processing as described below.

[0026] Usually, the PTFE film is a heat processable PTFE fluoropolymer however, any PTFE may be employed. The welding described above produces one or more heat affected zones which, left untreated may be prone to contribute to particle shedding. However, as described further herein, the heat affected zones and PTFE film generally may be treated to reduce such occurrence.

[0027] A heat affected zone is caused by heating the PTFE film to temperatures near or at, the melting point of the molecular components in the film. Generally, a heat affected zone is caused by heating the PTFE film to within about 15°C of the melting point of the PTFE film. Portions of the film tend to melt at different temperatures. For example, crystalline

portions of the film may melt at temperatures higher than the melting temperature associated with the PTFE film, while amorphous portions of the film may melt at temperatures lower than the melting temperature associated with the PTFE film. Thus, a heat affected zone may be created even if the melting temperature associated with the PTFE film is never actually reached. Additionally, in order to completely weld two pieces of PTFE film together, at least the melting point of the PTFE films should be reached and temperatures somewhat above the melting point may be beneficial. Heat affected zones are beneficially affected by the processes of the present invention, as is PTFE film without heat affected zones.

[0028] Generally, step 105 includes multiple welding steps, such that a complete PTFE liner is formed. As described above, a PTFE liner is usually a four-sided, two-dimensional bag with a spout called a fitment. The spout is also welded onto the liner. Thus, step 105 will generally include several welding steps.

[0029] With continued reference to Fig. 1, and as described above, in step 110, one or more PTFE films may be heat treated by placing the films at a predetermined temperature for a predetermined time period. In this manner, particle shedding and particle counts, as described above, may be minimized. There are a number of different temperatures and time periods that may be beneficial, but there are certain temperatures and time periods that show particular benefit. Particular techniques for determining an optimum temperature and time period are described in reference to FIGS. 2 through 10. Temperatures above the glass transition temperature of the material (about 130°C for many PTFE films) will begin to produce lower particulate shedding, as shown in more detail below. The heat treating process becomes particularly effective as temperatures exceeding 150°C are applied. As shown in more detail below, even higher temperatures are beneficial, until an optimal temperature is reached, but too high of a temperature can lead to increased particulate shedding as compared to the optimum temperature. Nonetheless, temperatures higher than the optimum temperature can be used if desired, as particle shedding may still be decreased with higher temperatures.

[0030] Generally, the predetermined temperature in step 110 is above about 150 degrees centigrade (C), more preferably above about 200°C, and most preferably about 228°C. Higher temperatures may also be used. It is preferable that temperatures be less than about 260°C. It should also be noted that a temperature range, such as 230°C ± 20°C, may be used. Additionally, films may be applied in a discontinuous or periodic manner. That is the PTFE material may be heated to the predetermined temperature and cooled to, for instance, room temperature, then reheated to the initial predetermined temperature. However, in order to

minimize total treatment time, the PTFE films may be continuously maintained or an alternate elevated temperature at the predetermined temperature for a particular time period, as reheating and cooling makes the total time required to heat treat PTFE films longer.

[0031] Concerning the predetermined time period, as shown in more detail below, a recommended time period is greater than about 20 hours, a more preferred time period is greater than 50 hours and a most preferred time period is about 100 hours. While shorter or longer time periods may be used, a time period of about 100 hours, as determined through the techniques given below, should provide the smallest amount of particulate shedding and the standard deviation thereof in the shortest time period. It should be noted that step 100 can take place after step 120, if desired.

[0032] Now that an overview of exemplary heat treatment techniques for PTFE films has been described, techniques for determining appropriate temperature and time periods for any particular PTFE film or, more likely, set of PTFE films will be described.

[0033] Referring now to FIG. 2, a dot plot of annealed test samples and controls is shown. This plot illustrates effects, in terms of particle count, of heat treating on PTFE liners. PTFE liners were filled with photoresist, inverted five times and counted at 0.2 microns 16 hrs after filling. The x-axis units are particles per milliliter (particles/ml) at 0.2 microns. The legend at the right of Fig. 2 contains a description of each set of samples tested. Each sample in a set of samples was a number of heat processable PTFE fluoropolymer films that were welded together into a liner. In this example, there are three PTFE liners per set. The welds were performed by using a welding machine that presses two PTFE films together, along a relatively thin line or set of lines, for a predetermined time at a predetermined temperature, where the predetermined temperature is equal to or greater than the melting point of the PTFE films. The "10 second cool" is a 10 second period of time used to cool the welded PTFE films to a predetermined cool temperature. Similarly, the "40 second cool" is a 40 second period of time used to cool the welded PTFE films to a predetermined cool temperature.

[0034] The samples, as shown in legend, were as follows: (1) a test set of PTFE liners with a 10-second cool followed by a 180°C heat treat for four days; (2) a test set of PTFE liners with a 40-second cool followed by a 180°C heat treat for four days; (3) a test set of PTFE liners with a 10-second cool followed by a 120°C heat treat for four days; (4) a test set of PTFE liners with a 40-second cool followed by a 120°C heat treat for four days; (5) a control set of PTFE liners with welds produced with a 10-second cool; (6) a test set of PTFE



liners with welds produced with 40-second cool; (7) a test set of PTFE liners with a 180°C heat treat for four days followed by a 10-second re-weld and a 40-second cool; (8) a test set of PTFE liners with a 180°C heat treat for four days followed by 40-second re-weld and a 10-second cool; (9) a test set of PTFE liners with a 120°C heat treat for four days followed by a 10-second re-weld and a 40-second cool; and (10) a test set of PTFE liners with a 120°C heat treat for four days followed by 40-second re-weld and a 10-second cool.

[0035] The dots plotted for each set of PTFE liners in FIG. 2 are allotted their own row in the figure. For instance, the dots plotted for the set entitled "10 second cool, 180° anneal" are shown on the row nearest the axis for particle count. Similarly, the dots plotted for the set entitled "40 second cool, 180° anneal" are shown on the second row from the axis for particle count.

[0036] FIG. 2 shows, among other things, that the effect of the amount of cooling time after welding is minimal, and the 180°C anneal produces PTFE liners having the lowest particle count for the temperatures selected.

[0037] Because the amount of agitation (i.e., inverting a filled PTFE liner five times) used in FIG. 2 is not typical in quality control testing, and because particle counts are generally taken using distilled water, samples were prepared to measure the effects of heat treatment on PTFE liners that were tested in DI water with one and five inversions. The heat treatment was performed at 180°C for five days. The sample PTFE liners were filled and particle tested with one hour of filling at 0.2 microns. The results are shown in FIG. 3, which is a dot plot of distilled water testing of control and annealed liners. FIG. 3 shows, among other things, that the annealed PTFE liners cause smaller particle counts than the corresponding control PTFE liners.

[0038] To determine the optimum settings for liner heat treatment, it is beneficial to select PTFE liners from a number of lots of liners. PTFE liners from each liner lot can then be heated for different time periods and for different temperatures. Thus, "sets" can be determined, where each set has a number of PTFE liners from certain lots, a selected temperature and a selected time period. The particle counts, for a chosen number of inversions of the PTFE liners, are then determined for the sets of liners and a regression analysis on the particle counts will determine an "optimal" temperature and "optimal" time period. The optimal values are theoretical and may differ slightly from an actual "optimal" temperature or time period. An example of this "optimization" technique follows.

[0039] Sample PTFE liners were prepared from three lots of liners. Each liner lot was heated for 20, 95, or 170 hours at 180°C, 220°C, or 260°C. Sample PTFE liners were filled with photoresist, inverted five times and particle counted at 0.2 microns at 16 hrs after filling.

[0040] Based on the results of the optimization technique for the three lots of liners tested, it appears that liner lot and heat treating temperature are the biggest contributors to particle levels and particle level variability, as indicated by the standard deviation. It appears that the relationships between liner lot, heat treating temperature, particle levels and standard deviation thereof are nonlinear with several second order interactions. If time is held constant at 20 hours, the liner lot and temperature curves (for particle count) look like what is shown in FIG. 4.

[0041] FIG. 4 is a calculated plot, based on actual data, showing how a particle count varies with heat treatment temperature for a number of liners held at a selected temperature for 20 hours. In the example of FIG. 4, actual data from different temperatures and liner lots were determined and placed into a program that used the data to determine the plot shown in FIG. 4. Thus, the plot shown in FIG. 4 is calculated based on actual data.

[0042] Regarding the temperatures shown in FIG. 4, the temperatures are as follows: -1 is equivalent to 180°C; 0 is equivalent to 220°C; and 1 is equivalent to 260°C. The response value is the particle count, in particles/ml. Fifteen PTFE liners were used from three different liner lots (lots 1, 0, and -1 in the figure). Thus, the plot of FIG. 4 reaches a low at about 236°C (0.4 times 40 plus 220°C), although this varies somewhat with liner lot.

[0043] FIG. 5 is a calculated plot, based on actual data, showing how a standard deviation of a particle count varies with heat treatment temperature for the liners used in FIG. 4. The temperatures, response values, and liner lots are the same as in FIG. 4. As with FIG. 4, the plot in FIG. 5 is calculated using actual data, and the data is the same used in FIG. 4. FIG. 5 shows that standard deviation of particle count is affected by heat treatment and reaches a low at about 244°C (0.6 times 40 plus 220°C), depending on liner lot.

[0044] FIG. 6 is a calculated plot showing how a particle count varies with heat treatment temperature for a number of liners held at a selected temperature for 95 hours. FIG. 6 is a plot generated through the same techniques used in regard to FIG. 4: fifteen PTFE liners, five from three different lots, were used and the resultant data from different temperatures and liner lots were determined and placed into a program that used the data to determine the plot shown in FIG. 6. FIG. 6 shows that particle count varies with temperature and liner lot.

[0045] FIG. 7 is a calculated plot showing how a standard deviation of a particle count varies with heat treatment temperature for the liners used in FIG. 6. FIG. 7 shows that the standard deviation of particle count varies with temperature and liner lot.

[0046] FIG. 8 is a plot calculated through techniques used to determine FIGS. 4 and 6 but based on a number of liners held at a selected temperature for 170 hours. It should be noted that the negative values of particle count are calculated and not observed. FIG. 9 is a plot calculated through techniques used to determine FIGS. 5 and 7 but calculated using the liners of FIG. 8. Both FIGS. 8 and 9 show that particle count and the standard deviation thereof, respectively, vary with temperature and liner lot.

[0047] Thus, FIGS. 4 through 9 show that particle count and the standard deviation thereof, respectively, vary with temperature and liner lot. In order to determine an optimum temperature and time period, a regression analysis is used on the determined data. The regression analysis of the data used to determine FIGS. 4 through 9 indicates that, to minimize particle levels and standard deviation at 0.2 microns, the temperature should be 228C for a time period of 100 hrs.

[0048] These values were used for a confirmation experiment where three lots of liners were heated treated for 100 hrs at 228°C. The results of the testing (five inversions in photoresist, 16hr rest with particle counting) are shown in FIG. 10. FIG. 10 is a dot plot of particle counts caused by heat treated PTFE liners. As can be seen by FIG. 10, the particle count and standard deviation thereof are small for heat treated PTFE liners. Thus, the present invention can reduce particle counts from about  $250 \pm 100$  particles/ml to about  $8 \pm 2$  particles/ml.

[0049] Note that liner lot variability, in terms of the standard deviation of the particle count, is also reduced. While there is still some variability, as the third liner lot has a few sample PTFE liners with particle counts greater than 10, the variability has been markedly decreased.

[0050] It is to be understood that the embodiments and variations shown and described herein are merely illustrative of the principles of this invention and that various modifications may be implemented by those skilled in the art without departing from the scope and spirit of the invention. In addition, the various assumptions made herein are for the purposes of simplicity and clarity of illustration, and should not be construed as requirements of the present invention.

CLAIMS

1. A method comprising:  
heating a polytetrafluoroethylene material to an elevated temperature; and  
maintaining said heating for a time sufficient to substantially reduce a  
particle count character of the polytetra fluoroethylene material.
2. The method of claim 1 further comprising applying a melting temperature to  
a portion of the polytetrafluoroethylene material for welding thereof prior to  
said heating.
3. The method of claim 2 wherein the melting temperature is within about  
15°C of a melting point of the polytetrafluoroethylene material.
4. The method of claim 2 wherein said applying forms a heat affected zone of  
the portion, said heating and said maintaining to affect the heat affected  
zone.
5. The method of claim 1 wherein the elevated temperature is above a glass  
transition temperature of the polytrafluoroethylene material.
6. The method of claim 1 wherein the elevated temperature is between about  
130°C and about 260°C.
7. The method of claim 1 wherein the time is between about 20 hours and  
about 100 hours.
8. The method of claim 1 wherein said maintaining occurs in a periodic  
manner comprising:  
cooling the polytetraethylene material; and reheating the  
polytetraethylene material.

9. The method of claim 1 wherein said maintaining occurs in a periodic manner comprising:  
cooling the polytetraethylene material; and  
reheating the polytetraethylene material.
10. A method comprising heating a polytetrafluoroethylene material to about 228° to substantially, reduce a particle count character thereof.
11. The method of claim 10 wherein said heating is for about 100 hours.
12. A polytetrafluoroethylene material having a particle count character reduced by application of an elevated temperature thereto.
13. The polytetrafluoroethylene material of claim 12 wherein the elevated temperature is between about 130° and about 260°C.
14. The polytetrafluoroethylene material of claim 12 wherein the application of the elevated temperature is for between about 20 hours and about 100 hours.
15. The polytetrafluoroethylene material of claim 12 wherein the elevated temperature is about 228°C and the application is for about 100 hours.
16. The polytetrafluoroethylene material of claim 12 to form a film for contracting a substance.
17. The polytetrafluoroethylene material of claim 16 wherein the substance is one of a liquid and a powder.
18. The polytetrafluoroethylene material of claim 17 wherein the film is for a package to contain the substance.

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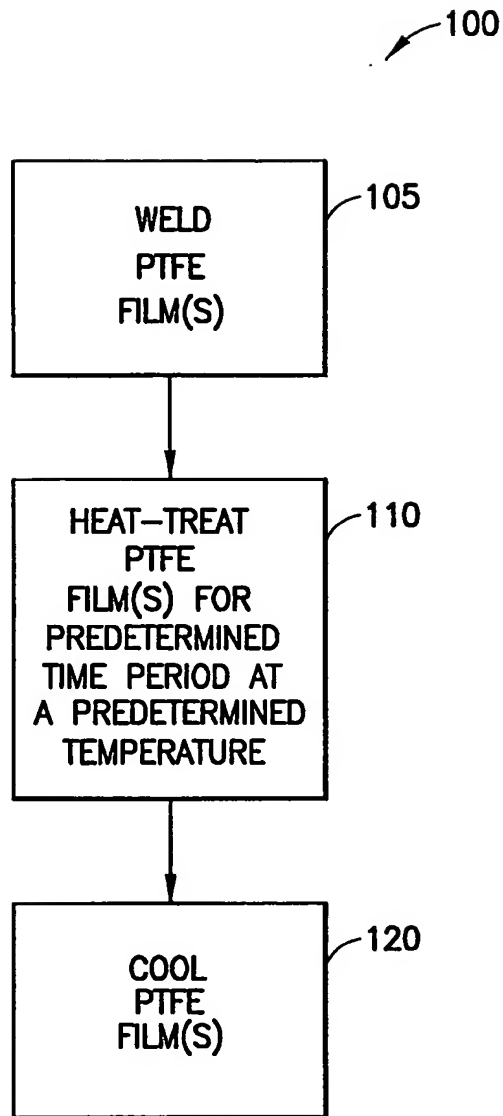
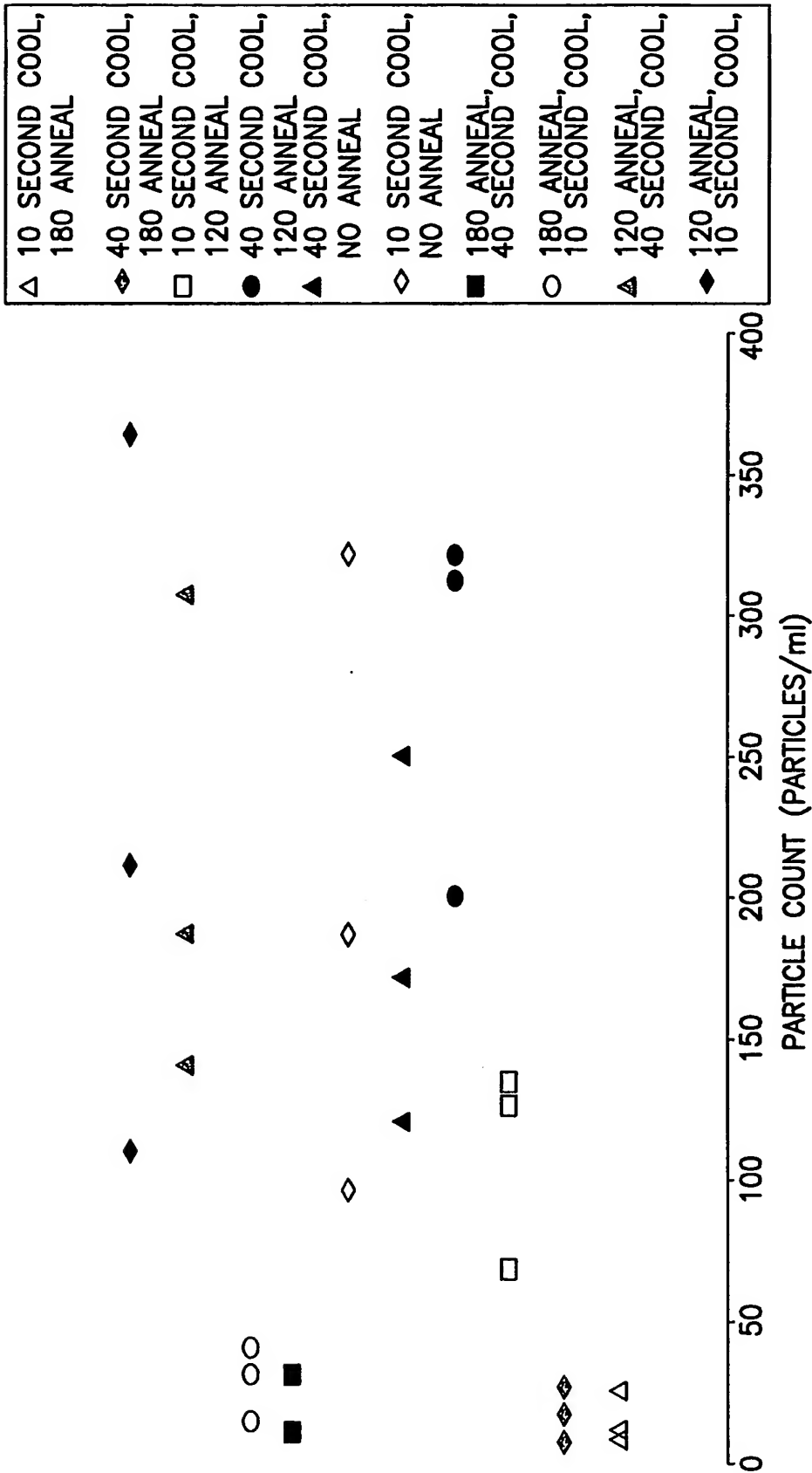


FIG.1

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DOT PLOT OF ANNEALED TEST SAMPLES AND CONTROL (NO ANNEAL)

FIG.2

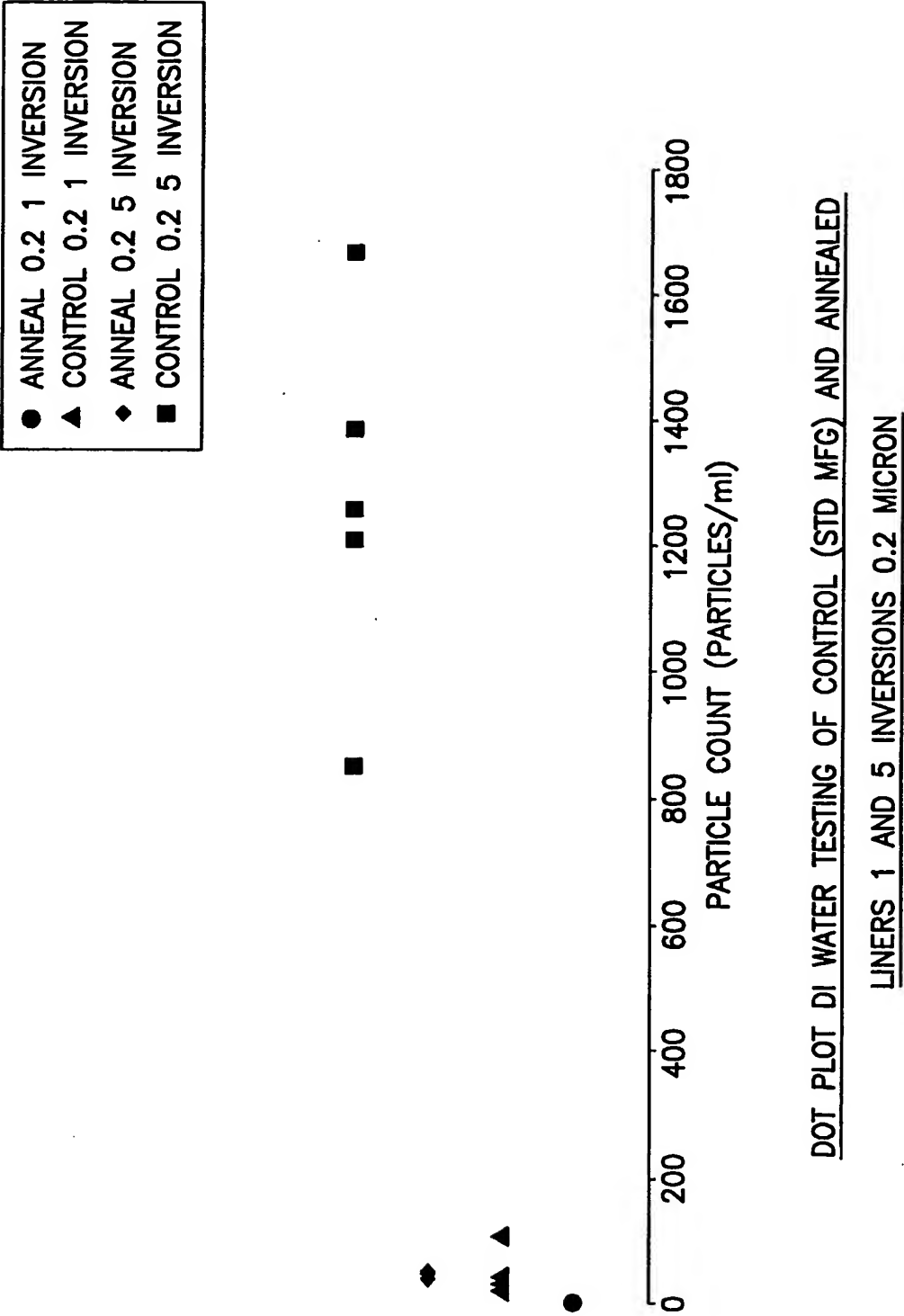
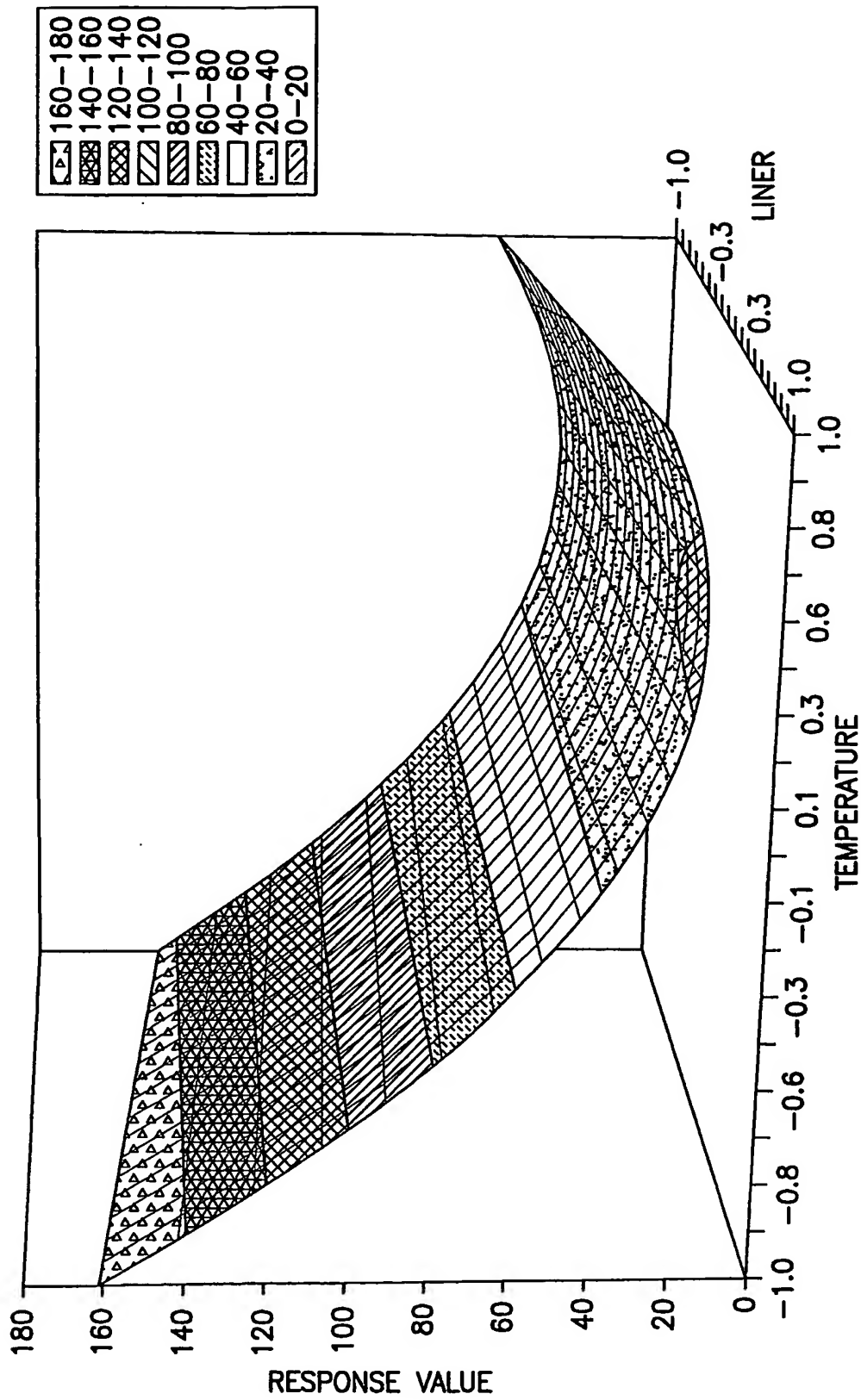


FIG.3



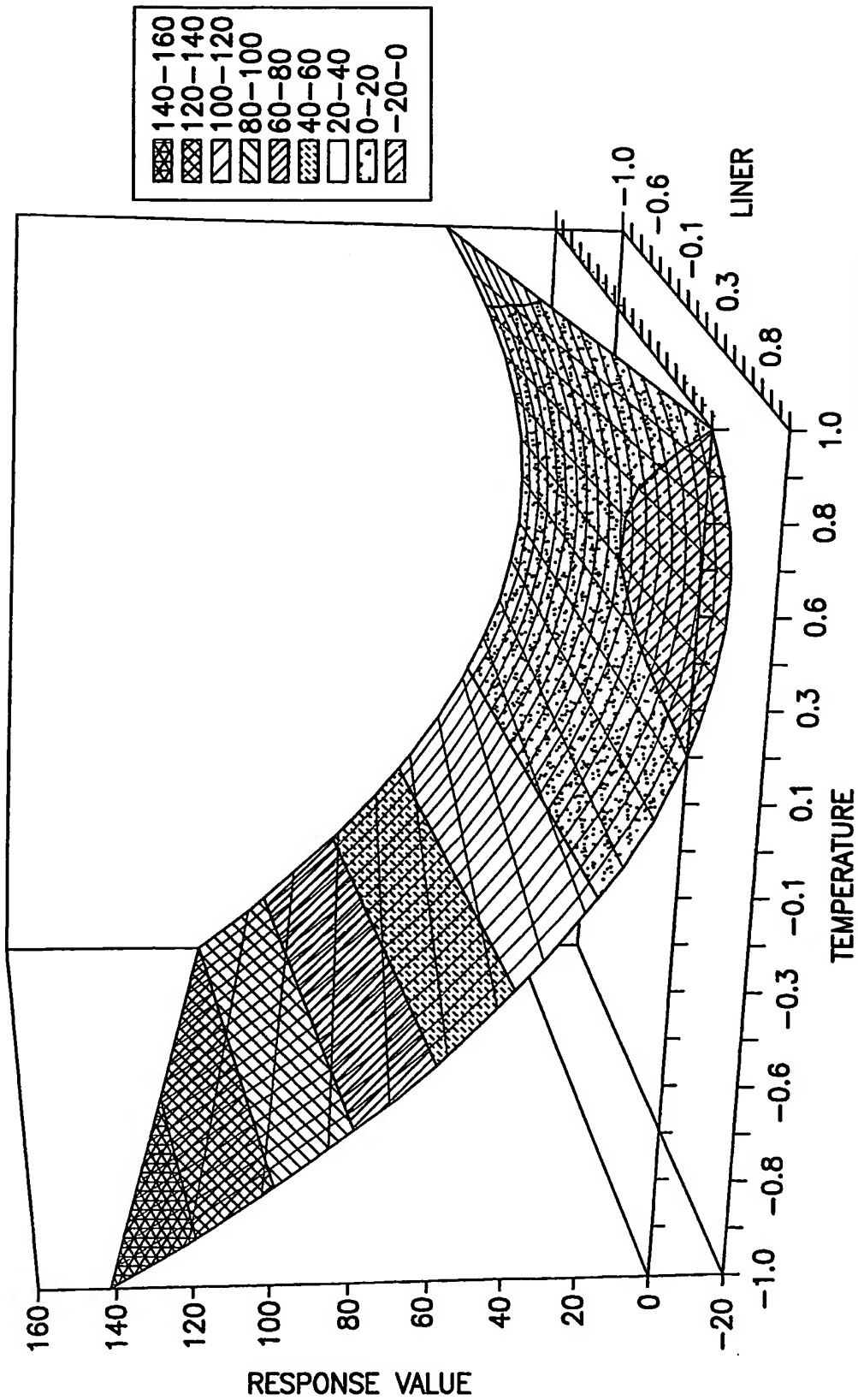
4/10



Y-HAT SURFACE PLOT OF (LPC AT 0.2 micron) A vs B CONSTANTS: C=-1

**FIG.4**

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S-HAT SURFACE PLOT OF (LPC AT 0.2 micron) A vs B CONSTANTS: C=-1

**FIG.5**

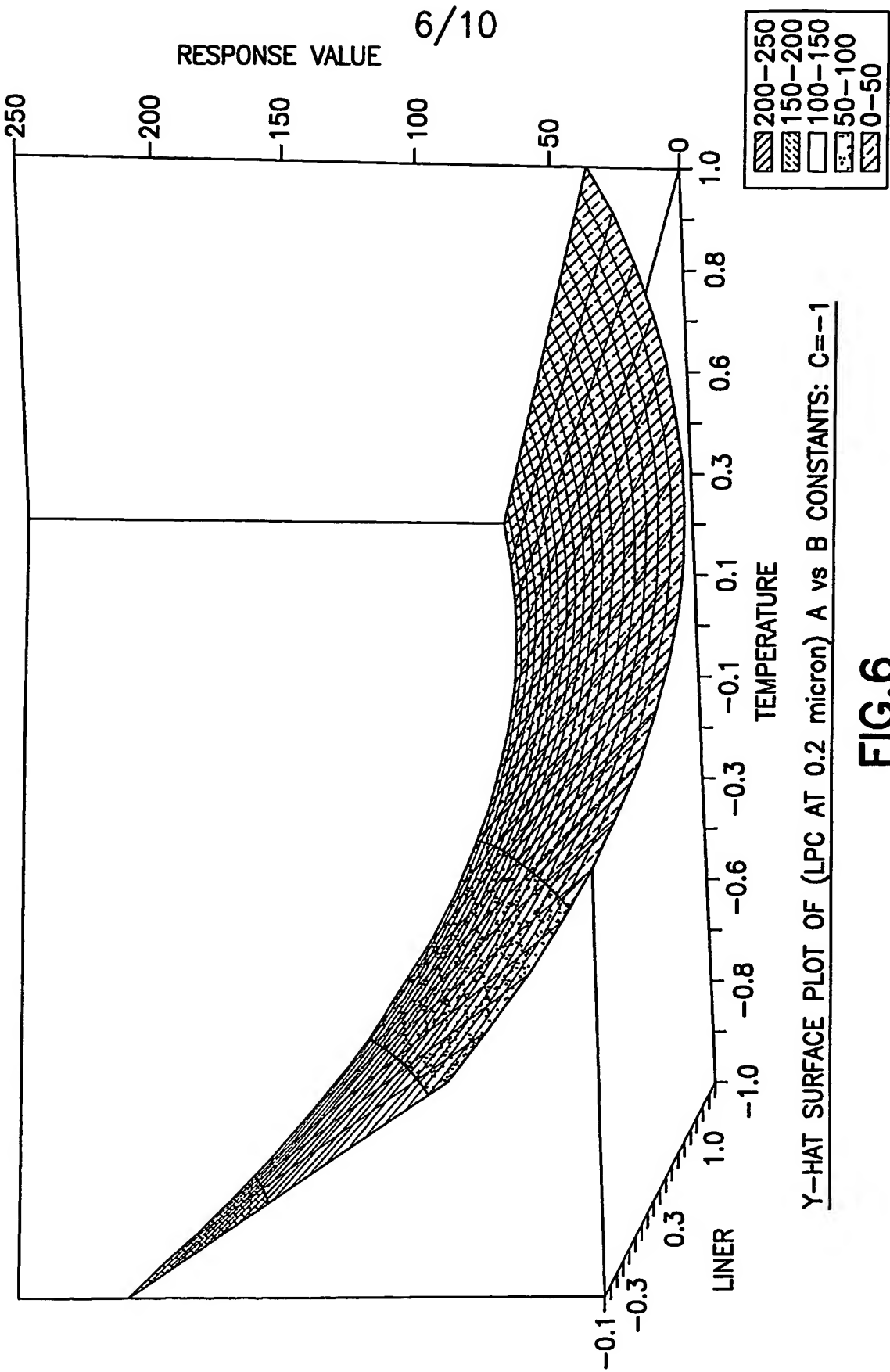


FIG.6

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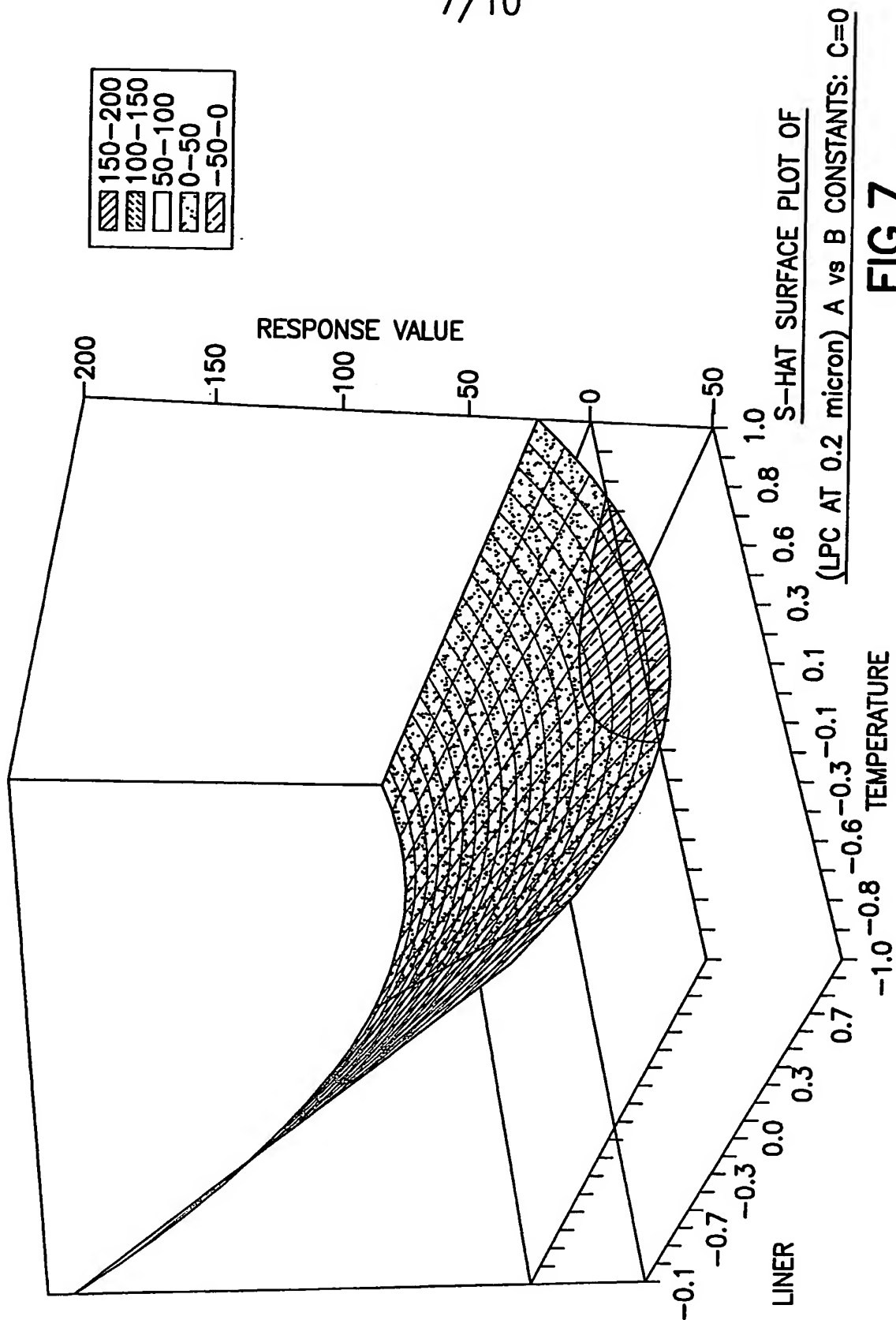
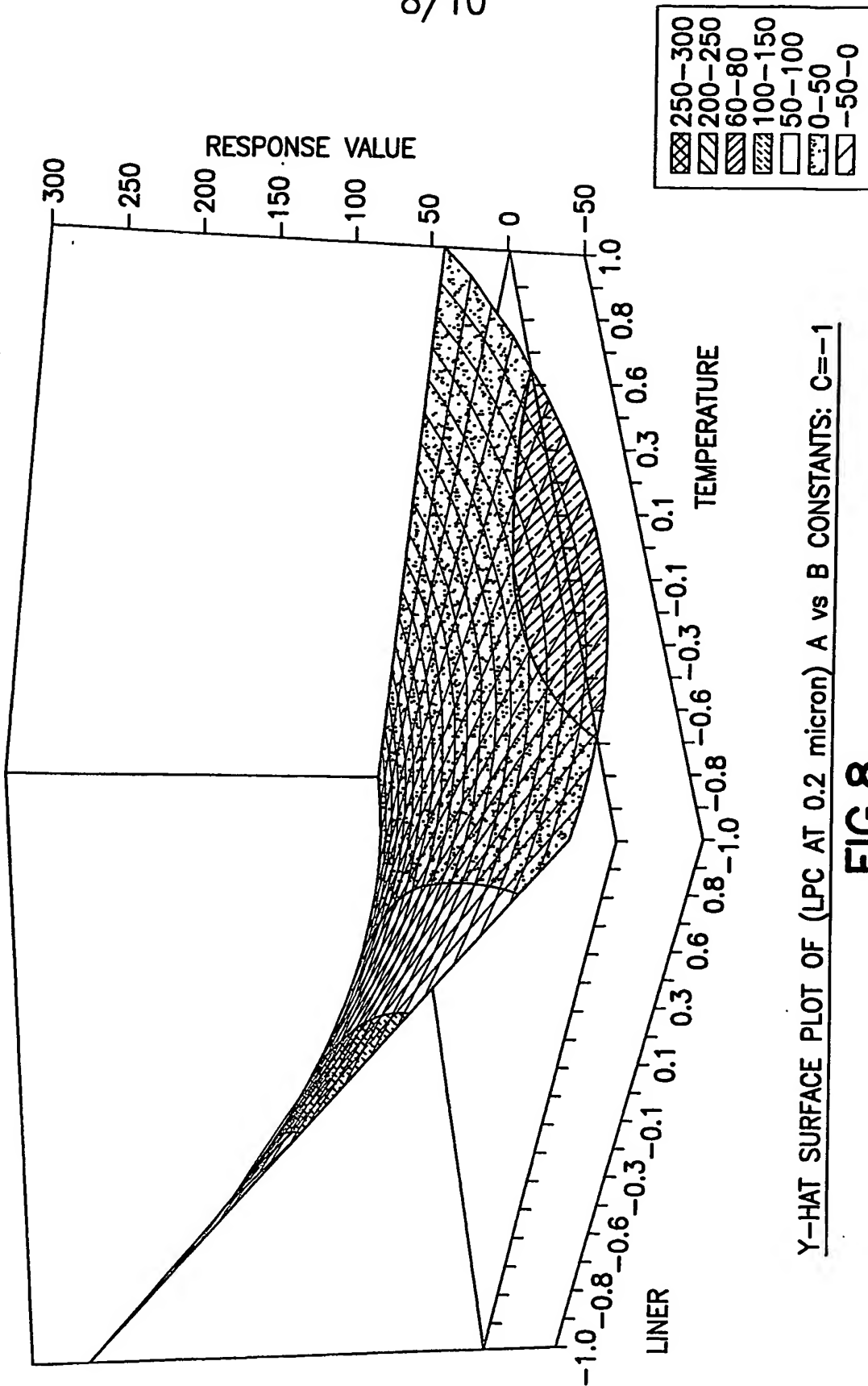


FIG. 7

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Y-HAT SURFACE PLOT OF (LPC AT 0.2 micron) A vs B CONSTANTS: C=-1

FIG.8

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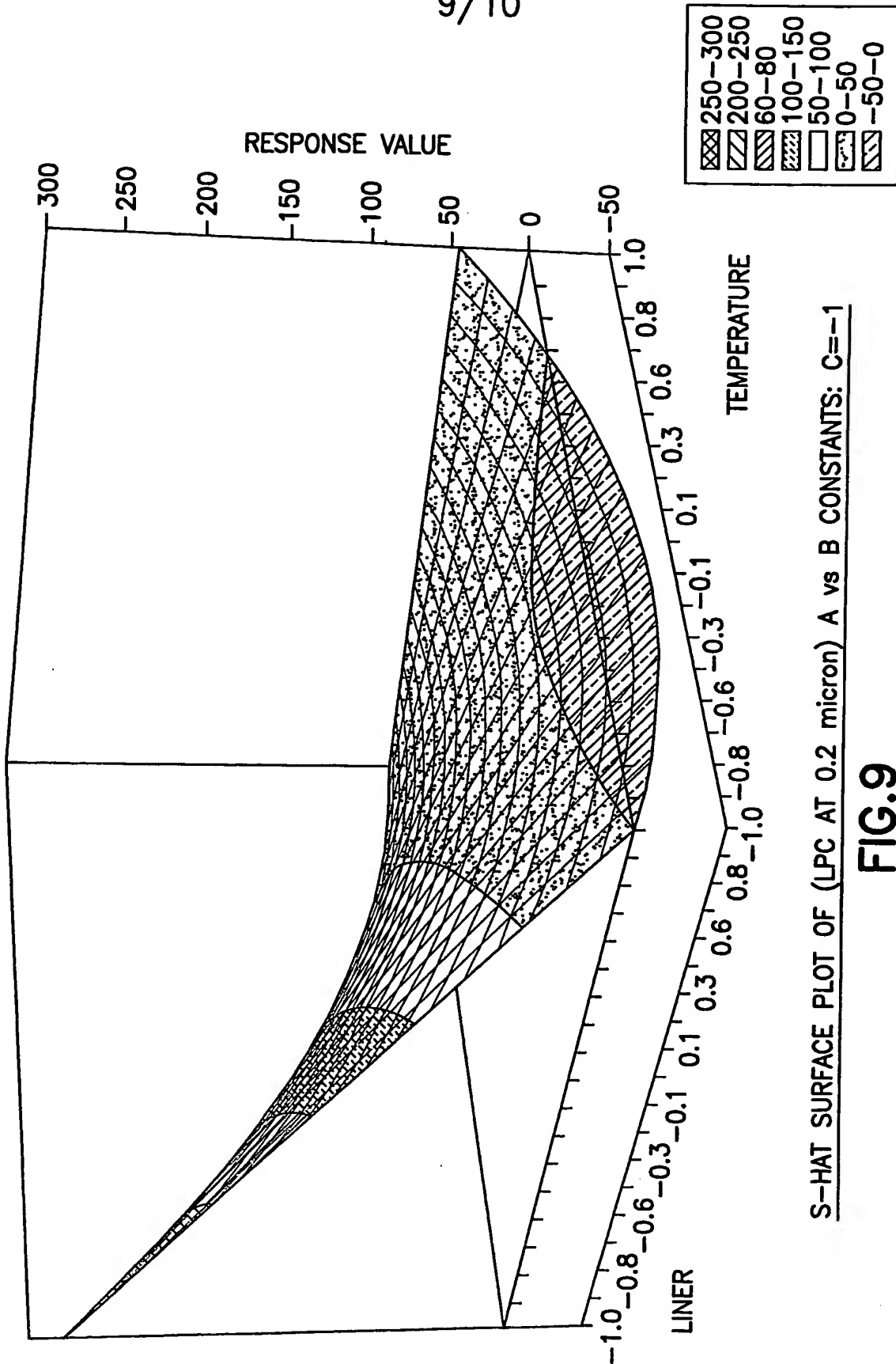
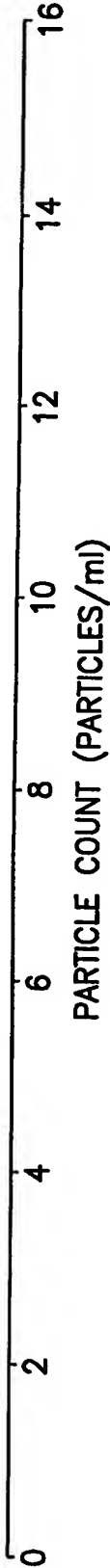
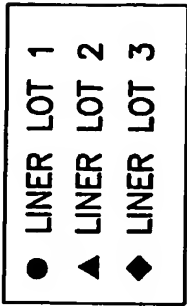


FIG.9

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DOT PLOT OF PARTICLE COUNTS CAUSED BY HEAT-TREATED PTFE LINERS

**FIG.10**

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/20024

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : C08F 6/00  
US CL : 528/503; 264/115

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 528/503; 264/115; 264/45.5; 528/481; 528/501

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

USPAT, USPG-PUB, EPO, JPO, DERWENT

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5,377,708 A (BERGMAN et al) 3 January 1995 (03.01.1995), abstract, line 16-20; column 10, line 62 - column 11, line 22; column 16, line 14-18; column 17, line 21-22; column 19, line 42-46; column 19, line 47-68; column 19, line 55-60; column 16, line 14-18; column 17, line 21-22.	1-18
X	US 3,432,511 A (REILING) 11 March 1969 (11.03.1969), column 1, line 15-17; column 2, lines 26-38 and 44-54.	1-18
X	US 4,220,511 A (DERBYSHIRE) 2 September 1980 (02.09.1980), abstract, line 1-12; column 1, line 32-58; column 3, line 11-25; column 7, line 47-54; column 3, line 26-30.	1-18

☐ Further documents are listed in the continuation of Box C.

☐ See patent family annex.

### \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

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later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

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document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y"

document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&"

document member of the same patent family

Date of the actual completion of the international search

30 September 2004 (30.09.2004)

Date of mailing of the international search report

21 OCT 2004

Name and mailing address of the ISA/US

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